

Unified Theoretical Framework for Polycrystalline Pattern Evolution

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The rate of curvature-driven grain growth in polycrystalline materials is well-known to be limited by interface dissipation. We show analytically and by simulations that, for systems forming modulated phases or non-equilibrium patterns with crystal ordering, growth is limited by bulk dissipation associated with lattice translation, which dramatically slows down grain coarsening. We also show that bulk dissipation is reduced by thermal noise so that those systems exhibit faster coarsening behavior dominated by interface dissipation for high Peierls barrier and high noise. Those results provide a unified theoretical framework for understanding and modeling polycrystalline pattern evolution in diverse systems over a broad range of length and time scales.

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Polycrystalline patterns are observed in very diverse systems including crystalline solids [1], colloidal systems [2, 3], various spatially modulated phases of macromolecular systems such as diblock copolymers [4, 5], and non-equilibrium (NE) dissipative structures [6]. When grain boundaries (GBs) between domains of different crystal orientation are mobile, those patterns generally coarsen in time to reduce GB length or area by elimination of smaller grains. This coarsening behavior has been extensively studied because of its practical importance for engineering polycrystalline materials [7] and its fundamental relevance for our general understanding of nonequilibrium ordering phenomena.

The ordering dynamics of modulated phases and NE patterns has been extensively studied theoretically [8–16] in the framework of model equations of the form

$$p\partial_t^2\psi + \alpha\partial_t\psi = -(-\nabla^2)^n\frac{\delta\mathcal{F}}{\delta\psi} + \eta, \quad (p, n = 0 \text{ or } 1), \quad (1)$$

where ψ is an order parameter appropriate to each system that can be globally conserved ($n = 1$) or non-conserved ($n = 0$), η is a noise uncorrelated in space and time with a variance determined by the fluctuation-dissipation relation $\langle\eta(\vec{r}, t)\eta(\vec{r}', t')\rangle = 2\alpha T(-\nabla^2)^n\delta(\vec{r}-\vec{r}')\delta(t-t')$, and \mathcal{F} is a Lyapounov functional with a minimum in a lattice ordered state. Eq. (1) has also been proposed as a theoretical framework—the phase-field-crystal (PFC) model—to study polycrystalline materials on diffusive time scales with ψ representing the crystal density field [17, 18]. While Eq. (1) has been traditionally studied for purely relaxational ($p = 0$) dynamics [8–17], propagative ($p = 1$) wave-like dynamics has also been introduced in the PFC framework to mimic phonon-mediated relaxation of the strain field [18].

Extensive computational studies of Eq. (1) have shown that the characteristic domain or grain size for both roll patterns [9–12] and hexagonal lattices [13–16] grows $\sim t^q$. The exponent q is typically much smaller than the $q = 1/2$ value expected for “normal grain growth” in

polycrystalline materials [19], and depends on parameters and noise strength [15, 16]. While there have been theoretical attempts to explain those exponents for roll patterns [9–12], the origin of this sluggish (low q) coarsening kinetics is still poorly understood, especially for lattices that relate to polycrystalline materials.

In this letter, we show that the sluggish ordering dynamics of crystal lattices results from the subtle effect of “bulk” dissipation linked to lattice translation, absent in crystalline solids, but present in Eq. (1) that lacks Galilean invariance. Internal stresses generated by GB motion are known to deform grains and even rotate them [21–24] during growth. Our main finding is that the associated dissipation can be, surprisingly, the dominant rate limiting mechanism of grain growth for a broad class of systems described by Eq. (1). This effect is dominant in the limit where Peierls-Nabarro (PN) barriers to dislocation motion are small and curvature-driven growth does not require thermal activation. This is the physically relevant limit for lattice patterns with a wavelength much larger than the atomic scale, which coarsen near zero T . Interestingly, in the opposite limit of high PN barrier and high noise, bulk dissipation plays a subdominant role because noise promotes modes of GB motion that reduce grain deformation. We summarize here our main results and will give a longer exposition elsewhere.

To highlight the effect of bulk dissipation, we first treat analytically the dynamics of a circular grain of radius $R(t)$ and misorientation $\theta(t)$ with respect to its surrounding single crystal matrix. A theory for this problem has been developed by Cahn and Taylor [22] for solids, and basic geometrical aspects of this theory have been validated by MD [24] and PFC [23] simulations. For small initial misorientation $\theta(0)$, grain rotation is geometrically necessary under the assumption that the number of dislocation along the GB is conserved. Since there are $n_d = 2\pi R\theta/b$ dislocations of Burgers vector magnitude b , this conservation condition implies that $R(\theta)\theta(t) = R(0)\theta(0)$. This rotation can also be inter-

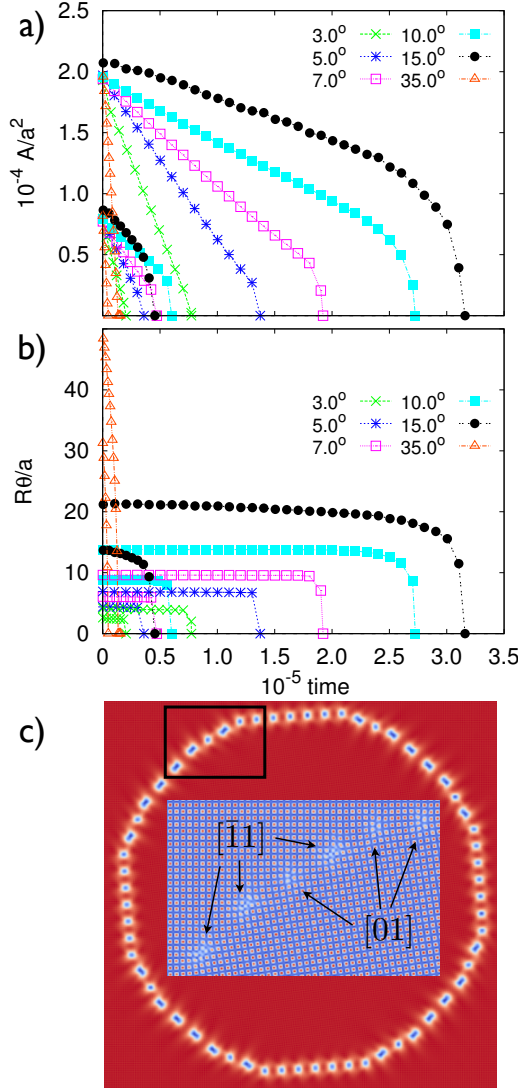


Figure 1: Results of isolated grain shrinkage simulations for $\epsilon = 0.12$ and $T = 0$ showing normalized grain area (a) and $R\theta/a$ (b) versus time for different initial grain areas and misorientations indicated in degrees. Grain rotation is present (absent) for misorientation less (larger) than $\sim 15^\circ$. (c) snapshot of defect field ϕ (see text) for the largest area 7° simulation showing the GB structure, with $\langle 11 \rangle$ ($\langle 10 \rangle$) dislocation cores appearing as elongated (circular) blue patches, and the corresponding ψ field inside the square region (inset).

interpreted as a consequence of the geometrical coupling between GB motion and a shear stress [22, 25, 26]. To treat dissipation, we focus for simplicity on non-converted dynamics ($n = 0$) but the results hold generally for any n or p . For $n = 0$ and $T = 0$, Eq. 1 implies

$$\dot{\mathcal{F}} + \dot{E}_k = - \int d\vec{r} \alpha (\partial_t \psi)^2, \quad (2)$$

where the dot denotes total derivative with respect to time and $E_k \equiv \int d\vec{r} (\partial_t \psi)^2 / 2$ is the kinetic energy of the ψ field. $\dot{\mathcal{F}}$ represents the rate of change of the

total GB energy of the circular grain, and thus $\dot{\mathcal{F}} = d[2\pi R(t)\gamma(\theta(t))]/dt$, where $\gamma(\theta)$ is the energy per unit length of GB. We write the total energy dissipation rate as the sum of interface and bulk contributions by splitting the integral on the right-hand-side (r.h.s.) of Eq. (2) as the sum $\dot{\mathcal{F}} = \dot{\mathcal{F}}_I + \dot{\mathcal{F}}_B$, where $\dot{\mathcal{F}}_I$ is over a thin annulus comprising the GB of thickness proportional to the dislocation core radius, and $\dot{\mathcal{F}}_B$ is over the bulk area of the inner grain. Dislocations move radially inward by pure glide at a velocity \dot{R} , and hence $|\partial_t \psi| \sim a^{-1} \dot{R}$ over an area a^2 around each dislocation, where a is the lattice spacing. Therefore, the total interface dissipation $\dot{\mathcal{F}}_I d\vec{r} \alpha (\partial_t \psi)^2 = n_d a^2 \alpha (a^{-1} \dot{R})^2 / m = \alpha 2\pi R \dot{R}^2 \theta / (mb)$, where m is an $O(1)$ dimensionless prefactor. This yields the expression for the mobility $M(\theta) = mb/(\alpha\theta)$ for $\theta \ll 1$ in agreement with previous studies [22, 26].

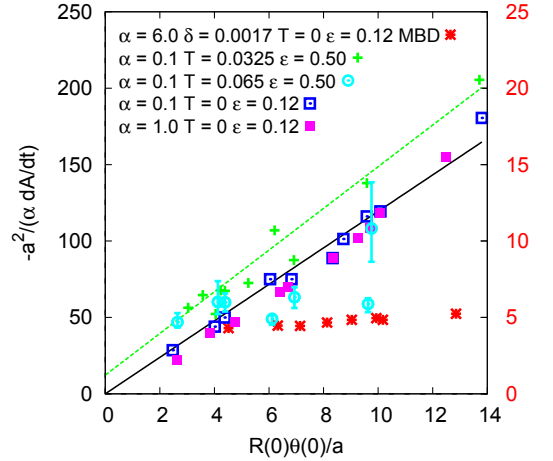


Figure 2: Comparison of theory (Eq. 4 for the octagonal grain with two dislocation types) shown as a black solid (green dashed) line for $\epsilon = 0.12$ ($\epsilon = 0.5$), and simulations of Eq. (1) (symbols). Red star symbols are for the modification of Eq. (1) with minimized bulk dissipation (MBD), which causes the area decrease rate to become independent of grain size and misorientation for small misorientation.

Next, to compute the bulk dissipation rate $\int_B d\vec{r} \alpha (\partial_t \psi)^2$, we compute the dissipation rate per unit area of a crystal field in uniform translation at velocity \vec{v} and then integrate the result over the entire grain area. In a region in uniform translation, $\psi(\vec{r}, t) \approx \psi_0(\vec{r} - \vec{v}t)$, where $\psi_0(\vec{r})$ is the equilibrium ψ -field that minimizes \mathcal{F} , and hence $\partial_t \psi = -\vec{v} \cdot \vec{\nabla} \psi_0$. The dissipation rate per area of crystal can therefore be written in the form $\alpha K v^2 / A_{u.c.}$ where $K \equiv \int_{u.c.} d\vec{r} (\vec{v} \cdot \vec{\nabla} \psi_0)^2$ is computed over the area $A_{u.c.}$ of one unit cell (u.c.). For a square lattice, $A_{u.c.} = a^2$ and K is independent of the direction of \vec{v} relative to the crystal axes, and reduces to $K = \int_0^a \int_0^a dx' dy' (\partial_{x'} \psi_0)^2$ where x' and y' are the principal crystal axes. Since $v = r\dot{\theta}$ in each

small region of a large rotating grain, where r is the radial polar coordinates, the total bulk dissipation rate is obtained by integrating $\alpha K v^2 / A_{u.c.}$ over the grain area: $\int_0^R 2\pi r dr \alpha K (r\dot{\theta})^2 / a^2 = \alpha \pi K R^4 \dot{\theta}^2 / (2a^2) = 2\alpha E_k$. Combining the above expressions for interface and bulk dissipation, Eq. (2) becomes

$$\dot{R}\gamma + R\gamma_{\theta}\dot{\theta} = -\alpha R\dot{R}^2\theta/(mb) - \alpha K R^4\dot{\theta}^2/(4a^2), \quad (3)$$

where we have neglected \dot{E}_k which can be shown to give a negligible higher order contribution (so that the $p = 0$ and $p = 1$ dynamics in Eq. (1) yield the same grain rotation dynamics). Finally, approximating the GB energy with a Read-Shockley law $\gamma(\theta) = E_0\theta(A_c - \ln \theta)$ valid for small θ , and using the condition $R(t)\theta(t) = R(0)\theta(0)$ that n_d is conserved, Eq. (3) yields a single dynamical equation for $R(t)$ that can be analytically solved. Its solution predicts that the grain area ($A = \pi R^2$) decreases linearly in time with a rate $\dot{A} = dA/dt$ given by

$$-a^2/(\alpha\dot{A}) = s_1 a^2/(mbE_0) + s_2 KR(0)\theta(0)/E_0, \quad (4)$$

where $s_1 = 1/(2\pi)$ and $s_2 = 1/(8\pi)$, and the first and second terms on the r.h.s. correspond to interface and bulk dissipation, respectively. Since $b \sim a$ and m and K are constants of order unity, Eq. (4) predicts that the ratio of bulk to interface dissipation is $\sim R(0)/a \gg 1$, implying that \dot{A} is entirely dominated by bulk dissipation, which holds for any lattice structure.

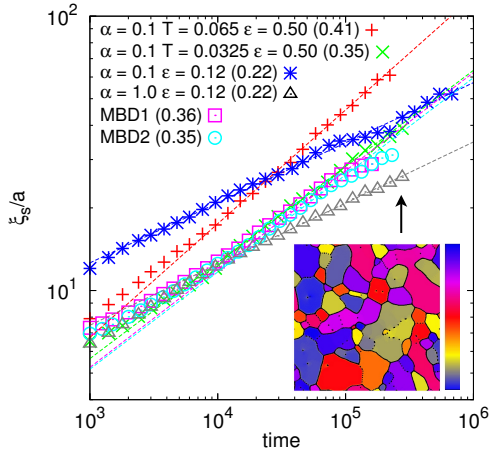


Figure 3: Plot of ordering length ξ_S (\sim mean grain size) vs. time from simulations of Eq. 1 (parameters in legends) and minimized bulk dissipation with $\epsilon = 0.12$, $\alpha = 8$, and $\delta = 3.1 \times 10^{-3}$ ($\delta = 6.3 \times 10^{-3}$) for MBD1 (MBD2); $T = 0$ when not given in legend. Numbers in parentheses are growth exponents from straight-line fits at large times (dashed lines). Inset: orientation field computed using a wavelet transform of ψ with a color bar ranging from -45° to 45° [31].

To test this prediction, we simulated Eq. (1) with $p = 1$ and $n = 0$ and $\mathcal{F} = \int d\vec{r} \omega$ where [27, 28]

$$\omega = \psi [-\epsilon + (\nabla^2 + 1)^2 (\nabla^2 + 2)^2] \psi / 2 + \psi^4 / 4 - \mu \psi, \quad (5)$$

has a stable square-lattice ground state that coexists with a metastable $\psi = 0$ liquid-like state for a range of $\mu < \mu_E$. This model was recently shown to produce GBs with a similar dislocation content as [001] tilt GBs in molecular dynamics (MD) simulations of face-centered-cubic bi-crystals [29]. In the analogy with a crystal-liquid system, \mathcal{F} represents the grand potential that is equal in both phases for an equilibrium value $\mu = \mu_E$ that depends on ϵ [28]. In the simulations reported here $\mu \approx 0.9\mu_E$, corresponding to $\mu = -0.9$ of $\epsilon = 0.12$ and $\mu = -1.67$ for $\epsilon = 0.5$. Since the height of the PN barrier scales $\sim \exp(-c/\epsilon^{1/2})$ where c is some constant (see, e.g. [12]), the height decreases rapidly with decreasing ϵ . For $\epsilon = 0.12$, this height is very small so that curvature-driven grain growth occurs at $T = 0$ even for $R \sim 100a$. In contrast, for $\epsilon = 0.5$ the barrier is large and even small grains are pinned. For $\epsilon = 0.5$, we carry out simulations at finite T and report results for two T values to highlight noise effects. Eq. (1) is simulated using a pseudo-spectral method described in [30] with more details in [31].

Fig. 1 shows plots of grain area and $R\theta/a$ versus time together with a snapshot that highlights the structure of the GB, consisting of dislocations with Burgers vectors described by the Miller indices $\langle 11 \rangle$ and $\langle 10 \rangle$. Accordingly, the grain is approximately shaped as an octagon with 4 facets made up of $[11]$, $[\bar{1}1]$, $[1\bar{1}]$, and $[\bar{1}\bar{1}]$ dislocations, and 4 others with $[10]$, $[01]$, $[\bar{1}0]$, and $[0\bar{1}]$ dislocations, respectively. Fig. 1a shows that \dot{A} is constant and depends on both initial grain size and misorientation, and that $R\theta$ is constant. Hence, the number of dislocations is conserved until dislocations become too closely spaced and annihilate during the final stage of grain collapse. In Fig. 2, we compare quantitatively \dot{A} values extracted from linear fits of A vs. time plots in the long linear regime before grain collapse to the predictions of Eq. (4) extended to an octagonal grain with two dislocation types: $E_0 = (E_0^{11} + E_0^{10})/2$ and $(mb)^{-1} = [(m_{10}b_{10})^{-1} + (m_{11}b_{11})^{-1}]/2$ together with the shape constants $s_1 = 1/(4\sqrt{2})$ and $s_2 = 0.0368$ related to the perimeter and area of an octagon [31]. This comparison shows an excellent quantitative agreement for $\epsilon = 0.12$ and $T = 0$ for two different α values, confirming that inertia ($p = 1$) is unimportant. The comparison for $\epsilon = 0.5$ with large PN barrier shows that bulk dissipation is still dominant for intermediate T ($T = 0.0325$). The slope of the curve predicted by Eq. (4) fits well the simulations results, but the curve has a finite intercept at the origin corresponding to a finite contribution of interface dissipation. This contribution is negligible for $\epsilon = 0.12$ where the intercept merges with the origin. For $\epsilon = 0.5$ and larger T ($T = 0.065$), bulk dissipation is reduced and the simulation data is no longer fitted by the theory. Analysis of dislocation dynamics in the simulations shows that this reduction results from thermally activated dislocation reactions. Those reactions reduce the number of dislocations, thereby allowing the grain to shrink with

less rotation and reducing the contribution of bulk dissipation relative to interface dissipation. Reduction of grain rotation by dislocation reaction is also observed in MD simulations of grain rotation [24].

To investigate the importance of bulk dissipation in grain growth, we simulated Eq. (1) with $p = 1$ and $n = 0$ in large systems of 512×512 unit cells with periodic boundary conditions. We also simulated a modified version of Eq. (1) that minimizes bulk dissipation (MBD) by the substitution $\alpha \rightarrow \alpha h(\phi)$ where $\phi(\vec{r}) = C \int d\vec{r}' \exp(-|\vec{r} - \vec{r}'|^2/2\zeta^2) |\nabla \psi(\vec{r}')|^2$ is a smoothly spatially varying crystalline order parameter (illustrated in Fig. 1b for a rotating grain), with $\zeta = a/2$ and the normalization constant C chosen such that $\phi = 1$ in ordered regions of the lattice and $\phi < 1$ in disordered regions (dislocation cores, GBs, etc). The function $h(\phi)$ given in [31] has limits $h(1) = \delta$ and $h(\phi) \sim O(1)$ in ordered and disordered regions, respectively, thereby allowing us to minimize the ratio of bulk to interface dissipation by choosing $\delta \ll 1$. Theory for MBD is identical to Eq. (4) with the second term on the r.h.s. multiplied by δ . Simulation results in Fig. 2 (red stars) confirm that bulk dissipation is negligible for $\delta \ll 1$.

We characterized the coarsening behavior by measuring the ordering scale ξ_S using the half width δk at half peak of the structure factor $S(k, t) = \langle \psi(\vec{k}, t) \psi(-\vec{k}, t) \rangle$ (fitted to a squared Lorentzian), where the angular brackets denote averages over all orientations of \vec{k} in the same simulation [10, 31]. Plots of $\xi_S \equiv \delta k^{-1}$ vs. time in Fig. 3 for simulations of Eq. (1) with $\epsilon = 0.12$ and $T = 0$ yield a small coarsening exponent $q \approx 0.22$ as in previous studies of hexagonal lattices [13–16]. However, MBD simulations yield a substantially larger exponent $q \approx 0.35$, thereby demonstrating that bulk dissipation associated with lattice translation has a significant influence on the coarsening kinetics even though grain rotation is more constrained in a polycrystal. This conclusion is corroborated by an analysis of the simulations, which shows that grain rotation is more common with MBD and by computing the ratio of interface and total dissipation. This ratio decreases to a small value with constant α but remains approximately unity with MBD [31]. Finally, simulations for $\epsilon = 0.5$ show that the growth exponent increases markedly with T , as expected from the reduction of bulk dissipation by dislocation reactions. More important here than the precise values of the exponents, which are not exactly constant in time, is the demonstration of the key role of bulk dissipation in the ordering dynamics. Aspects of the present results, in particular the theoretical prediction of the striking dependence of grain area shrinkage rate on initial grain size and misorientation (Eq. 4), should be testable experimentally in a wide range of lattice forming systems.

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